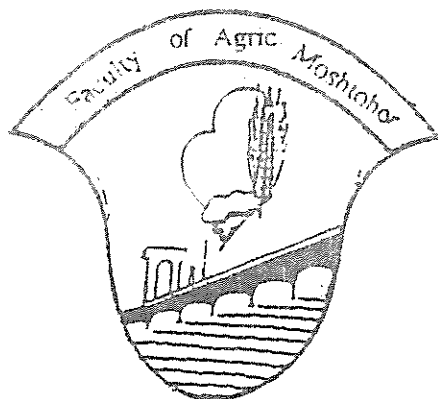


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**DETERMINATION OF CERTAIN ORGANOCHLORINE AND
ORGANOPHOSPHORUS PESTICIDE RESIDUES IN HOME-
PRODUCED TOMATOES AND CUCUMBERS USED FOR
CONSUMPTION IN TWO EGYPTIAN GOVERNORATES
BY**

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ABSTRACT

Residues of the organochlorine pesticides namely, (alpha + beta hexachlorocyclohexane, lindane, aldrin, dieldrin, endrin and total DDT) and the organophosphorus pesticides (pirimiphos-methyl and malathion) were determined in home-produced tomatoes and cucumbers used for consumption in two Egyptian Governorates.

The analysed samples were collected randomly and periodically during March 1994 - April 1995 from the markets in Kalyubia and Cairo Governorates.

Results showed that most of the tested samples contained very low amounts of alpha + beta-HCH, lindane and DDT, below the acceptable limits of the FAO. Aldrin, dieldrin and endrin residues were found frequently in a few number of the investigated samples, below the acceptable limits. Residues of alpha and beta HCH were found to be higher than the acceptable limit in, only, two samples of cucumbers. The organophosphorus insecticides pirimiphos-methyl and malathion were detected in most of the tomato and cucumber samples at very low levels below the allowed maximum residue limits.

INTRODUCTION

The organochlorine pesticides have been used extensively for pest control over the past 40 years. It has been established that this group persisted in the soil for long time, while organophosphorus pesticides are moderately persistent in soil. These pesticide residues find their ways from the soil to the plants contaminating the plant products consumed by man and animals. In Egypt, several investigations were conducted on residual behaviour of organophosphate pesticides in different field crops (Dogheim and Riskallah, 1984; Hegazy *et al.*, 1987; Monir and Dogheim, 1987; Hegazy *et al.*, 1988a & b and Abdallah *et al.*, 1993)

Tomatoes and cucumbers are widely grown vegetables in Egypt and have a number of pesticidal treatments for the control of insect pests and fungal diseases.

Monitoring the pesticide residues in those highly consumed vegetables is of great importance for human health. The aim of the present study was to detect the presence of organochlorine pesticides (alpha and beta HCH, lindane, dieldrin, endrin and DDT) and organophosphorus pesticides (pirimiphos-methyl and malathion) residues in tomato fruits and cucumbers used for consumption in Kalyubia and Cairo Governorates.

MATERIALS AND METHODS

Sampling:

Forty eight tomato fruits and cucumbers samples, each of half a kilogram were collected periodically during March 1994 - April 1995 from different markets in Kalyubia, Moshtohor, Takh, Benha and Kalyub) and Cairo (El-Dokki, Dokki, Shobra and Mataria), Governorates. Samples were kept at -20°C until analysis.

Samples were analysed for organochlorine pesticides, (HCH α and β , lindane, Aldrin, Dieldrin, Endrin and DDT) and the most persistent organophosphorus pesticides, (pirimiphos-methyl and malathion) residues, at the Laboratory of Pesticides, Plant Protection Dept. of the Faculty of Agriculture at Moshtohor, Zagazig University.

Extraction:

In this work the analytical procedure of El-Lakwah *et al.* (1989a & b) was performed. Half a kilogram of each sample was macerated for approximately 15 minutes. Twenty five grams of the macerate were transferred to 500 ml roundbottom flask, 50 ml of acetone and 25 gm of anhydrous sodium sulphate were added. Samples were shaken in Janke & Kunkel, KS 250, Shaker for 30 minutes. 10 ml of n-hexane was added and shaken further for 30 minutes. The samples were transferred to a funnel fitted with filter paper (Whatman No. 41, 15 cm). Fifty ml of the filtrate were transferred into 250 ml roundbottom flask containing 0.5 ml n-hexadecane. The sample size was reduced to about 1 ml using a rotary evaporator at 45 to 50°C.

Clean up:

Four grams celite was added into the same 250 ml flask. The content of

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The upper organic layer was drained into a chromatographic column provided with a plug of purified cotton, a mixture of (9.3 g Al_2O_3 + 0.7 g Fuellers earth) and 1 cm anhydrous sodium sulphate layer. The column was eluted by 40 ml of petroleum ether for chlorinated pesticides.

The eluate was collected in 250 ml round bottom flask containing 0.5 ml n-hexadecane and was evaporated. The remaining volume was transferred with 10 ml n-hexane in 10 ml volumetric flask. 1 μ l was used for GLC analysis.

For organophosphorus pesticides, the same column was eluted by 40 ml of a mixture of dichloromethane, acetone and benzol in ratio of 5 : 1 : 1. The eluate was collected, evaporated to dryness and transferred with 10 ml n-hexane. All solvents and chemicals used were analytical grades. Two μ l was used for GLC analysis.

Gas-liquid chromatography (GLC) analysis:

Hewlett packard series II gas chromatograph equipped with electron capture detector, and nitrogen phosphorus detector and provided with Hewlett packard HP 3396-Integrator was used for determination of organochlorine and organophosphorus insecticide residues.

The parameters were as follows: silanized pyrex glass column (1.8 m X 2mm ID) packed with 1.5% OV-17+1.95% OV 202 on Chromosorb WHP, 80/100 mesh.

For chlorinated insecticide analysis: column temperature 195°C, detector temperature 300°C, injector temperature 230°C, nitrogen (carrier gas) 25 ml/min.

For organophosphate analysis, column temperature 195°C, detector temperature 220°C, injector temperature 230°C, nitrogen carrier gas 25 ml/min., hydrogen 3-4 ml/min., air 100 ml/min.

Samples injections were alternated with injections of standards. Preliminary results from recovery studies yield 90-100% of chlorinated pesticides (Lindane, Alpha and Beta HCH, total DDT, Aldrin, Dieldrin and Endrin), and 80-90% of organophosphorus pesticides (malathion and pirimiphos-methyl).

RESULTS AND DISCUSSION

1): Pesticide residues in tomato fruits.

| Location | Organochlorine pesticides (ppm) | | | | | | Organophosphorus pesticides (ppm) | |
|----------|---------------------------------|---------|--------|----------|--------|-----------|-----------------------------------|-----------|
| | $\alpha + \beta$ HCH | Lindane | Aldrin | Dieldrin | Endrin | Total DDT | Pirimiphos methyl | Malathion |
| RL | 0.02 | 1.0 | 0.1 | 0.1 | 0.02 | 0.1 | 1.0 | 0.5 |
| Gov.: | | | | | | | | |
| hor | 0.001 | 0.001 | - | - | - | - | - | 0.02 |
| hor | 0.002 | 0.01 | - | - | 0.005 | 0.003 | - | 0.07 |
| hor | 0.001 | 0.03 | 0.001 | - | - | 0.006 | - | - |
| | - | 0.01 | - | - | 0.003 | 0.020 | - | 0.07 |
| | - | 0.01 | - | - | - | - | - | 0.08 |
| | 0.001 | 0.07 | - | - | - | - | 0.02 | - |
| | 0.005 | 0.02 | - | - | 0.003 | 0.040 | 0.03 | 0.16 |
| | 0.002 | 0.05 | - | - | - | - | 0.02 | - |
| | 0.003 | 0.02 | - | - | - | - | 0.02 | - |
| | 0.003 | 0.02 | - | - | - | 0.030 | 0.03 | 0.06 |
| b | 0.001 | 0.10 | 0.003 | - | - | 0.002 | 0.02 | 0.03 |
| b | 0.003 | 0.05 | - | - | - | - | - | - |
| b | 0.001 | 0.01 | - | - | - | 0.008 | - | - |
| ge* | 0.002 | 0.03 | 0.002 | - | 0.004 | 0.016 | 0.02 | 0.07 |
| Gov.: | | | | | | | | |
| polis | 0.007 | 0.08 | - | - | - | - | - | - |
| polis | 0.003 | 0.04 | - | 0.003 | - | - | 0.02 | 0.03 |
| polis | - | 0.01 | - | - | - | - | - | - |
| i | 0.001 | 0.08 | - | - | - | 0.020 | - | 0.10 |
| i | 0.003 | 0.09 | 0.002 | - | - | 0.010 | 0.03 | 0.04 |
| i | 0.001 | 0.10 | - | - | - | 0.030 | - | - |
| ra | 0.001 | 0.05 | - | - | - | 0.020 | - | 0.06 |
| ra | 0.002 | 0.06 | - | - | - | 0.010 | - | 0.08 |
| ra | 0.001 | 0.01 | - | - | - | 0.020 | - | - |
| ria | 0.003 | 0.03 | - | 0.002 | - | - | 0.02 | 0.02 |
| ria | 0.001 | 0.01 | - | - | - | - | - | 0.03 |
| ria | 0.003 | 0.08 | - | - | - | 0.008 | 0.02 | 0.04 |
| range* | 0.002 | 0.05 | 0.002 | 0.003 | - | 0.020 | 0.02 | 0.05 |

*B. Average were calculated considering only the samples that showed insecticidal residues.

Lindane residues ranged from 0.001-0.1 ppm in the tested samples in both Kalyubia and Cairo Governorates. These residue limits were below the maximum acceptable limit (1.0 ppm).

Aldrin residue was found only in 2 samples at Kalyubia, one from Moshtohor at value of 0.001 ppm, while the other sample was from Kalyub at value of 0.003 ppm. Aldrin residue was detected only in one sample at Cairo from Dokki market at value of 0.002 ppm. All samples were below the maximum residues limit (0.1 ppm).

No residues of dieldrin was found in all samples from Kalyubia, while in Cairo dieldrin residues were found in one sample from Heliopolis at 0.003 ppm and in another sample from Mataria market at level of 0.002 ppm. Both samples were under the acceptable limit (0.1 ppm).

In Case of endrin, the residues were detected in one sample from each of Moshtohor, Tikh and Benha markets at levels of 0.005, 0.003 and 0.003 ppm, respectively. All these values were under the acceptable limit (0.02 ppm). No residues of endrin was found in Cairo markets.

DDT residues ranged from 0.002-0.03 ppm in both Kalyubia and Cairo Governorates, in levels below the acceptable limit (0.1 ppm).

For organophosphorus insecticide residues of pirimiphos-methyl was 0.02-0.03 ppm in Kalyubia and Cairo, these levels were under the acceptable limit (1.0 ppm). Malathion residues ranged from 0.02-0.16 ppm in levels under the acceptable limit (0.5 ppm).

Organochlorine and organophosphorus pesticides residues in cucumbers are given in Table (2). Values were means of two analysed samples for each location. Alpha and beta HCH residues ranged from 0.001-0.06 ppm in both Kalyubia and Cairo Governorates. Values were mostly in levels under the acceptable limit (0.02 ppm), except for one sample at Kalyub market, where the value was 0.06 ppm. Also in Cairo, one of the samples at Mataria Market was equal to the acceptable limit while one sample at Dokki was found to be higher than this limit (0.06 ppm).

Lindane residues were found to be lower than the acceptable limit (1.0 ppm) and ranged from 0.01-0.14 ppm.

Pesticide residues in cucumbers.

| | Organochlorine pesticides (ppm) | | | | | | Organophosphorus pesticides (ppm) | |
|---|---------------------------------|---------|--------|----------|--------|-----------|-----------------------------------|-----------|
| | $\alpha+\beta$ HCH | Lindane | Aldrin | Dieldrin | Endrin | Total DDT | Pirimphos methyl | Malathion |
| | 0.02 | 1.0 | 0.1 | 0.1 | 0.02 | 0.1 | 1.0 | 0.5 |
| A | 0.003 | 0.10 | - | - | - | 0.003 | 0.02 | 0.03 |
| | - | 0.07 | - | - | - | - | - | - |
| | 0.002 | 0.03 | - | 0.003 | - | - | 0.03 | 0.06 |
| | 0.001 | 0.02 | - | - | - | - | - | - |
| | 0.001 | 0.04 | - | - | - | - | - | - |
| | - | 0.03 | - | - | - | - | 0.02 | - |
| | 0.003 | 0.09 | - | - | 0.003 | 0.004 | - | - |
| | - | 0.13 | - | - | - | 0.001 | 0.01 | - |
| | 0.003 | 0.05 | - | - | - | 0.002 | 0.04 | 0.04 |
| | 0.003 | 0.05 | - | - | - | 0.020 | 0.04 | 0.04 |
| | 0.060 | 0.01 | - | - | - | - | 0.08 | - |
| | 0.002 | 0.02 | - | - | - | - | - | - |
| | 0.011 | 0.05 | - | 0.003 | 0.003 | 0.006 | 0.03 | 0.04 |
| | 0.010 | 0.12 | - | - | - | 0.040 | 0.01 | - |
| B | 0.001 | 0.03 | 0.001 | - | - | 0.030 | 0.07 | 0.05 |
| | 0.002 | 0.07 | - | - | - | 0.004 | 0.01 | - |
| | 0.002 | 0.03 | - | - | - | 0.006 | 0.08 | 0.03 |
| | 0.004 | 0.05 | - | - | - | - | 0.03 | - |
| | 0.060 | 0.02 | - | - | 0.004 | 0.040 | 0.01 | 0.03 |
| | 0.003 | 0.05 | 0.001 | - | - | 0.030 | 0.10 | 0.08 |
| | 0.002 | 0.03 | - | - | - | 0.004 | - | - |
| | 0.001 | 0.01 | - | - | - | 0.006 | - | - |
| | 0.020 | 0.14 | - | - | - | 0.030 | 0.02 | - |
| | 0.003 | 0.04 | - | - | - | - | 0.01 | - |
| | 0.002 | 0.03 | - | 0.002 | - | 0.002 | 0.01 | - |
| | 0.001 | 0.01 | - | 0.002 | 0.004 | 0.019 | 0.04 | 0.05 |

In case of dieldrin, residues were found only in Moshtohor market 0.003 ppm and in Mataria market 0.002 ppm in levels under the acceptable limit 0.1 ppm

Endrin residues were found only in 2 samples, in Benha at level 0.003 ppm and in Dokki at level of 0.004 ppm, both samples were under the acceptable limit (0.02 ppm)

Total DDT residues were detected in all markets in levels ranged from 0.001-0.04 ppm under the acceptable limit (0.1 ppm) except Tikh market where no DDT residues was detected.

For organophosphorus insecticide residues, pirimphos-methyl was detected in levels 0.01-0.08 ppm under the acceptable limit (1.0 ppm).

Malathion residues ranged from 0.03-0.08 ppm in levels under the acceptable limit (0.5 ppm). No residues of malathion were detected in Tikh market and Mataria market.

The obtained results showed that most organochlorine and organophosphorus pesticide residues in tomatoes and cucumbers, were below the acceptable limit. Residues of alpha and beta HCH were found to be higher than the acceptable limit in very few samples of cucumbers.

Alpha and beta HCH and lindane were found in all samples, while aldrin, dieldrin, endrin and total DDT residues were detected only in some samples. These results may prove that they are still contaminating the soil in spite of discontinued use of these compounds. Residues of organochlorine insecticides could be absorbed by roots or leaves, metabolised and translocated in non insecticidal amounts, to leave persistent and undesirable residues at harvest. (Finlayson and Maccarthy, 1973). Therefore, the detected residues of the organochlorine pesticides in tomato fruits and cucumbers could be due to their persistence and accumulation in the soil and absorption by plant roots (Polizu and Floru, 1972).

Metcalf (1955) indicated that efficient translocation and movement of pesticide residues appear to be largely a function of the water solubility of the compound. The organochlorine pesticides DDT, aldrin, dieldrin, heptachlor were found to have a very low solubility in water.

The presence of organochlorine pesticides in the present results is expected to be due to the absorption of the residues by plant roots. While in case of the organophosphorus pesticide, Malathion, Metcalf (1955) showed that

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تقدير متبقيات بعض المبيدات الكلورونية والفوسفورية العضوية في الطماطم والخيار المنتجة محليا والمعروضة للاستهلاك في محافظتين مصريتين فارس أمين اللقوه ، محمد شوقي حامد ، أحمد عبدالغفار درويش على محمد شمس الدين قسم وقاية النبات كلية الزراعة بمشتهر - جامعة الزقازيق مصر .

أجريت هذه الدراسة بغرض تقدير متبقيات المبيدات الكلورونية (ألفا وبيتا هكساكلوروسيكلو هكسان، اللندان، الألدرين، ديالدرين، إندرين و د.د.ت) والمبيدات الفوسفورية العضوية (بريموفوس-ميثيل والملاثيون) في ثمار الطماطم والخيار المنتجة محليا والمعروضة للاستهلاك في محافظتين مصريتين.

وجمعت العينات التي تم تحليلها في هذه الدراسة بطريقة عشوائية من الأسواق بمحافظة القليوبية والقاهرة خلال المدة من مارس ١٩٩٤ وحتى أبريل ١٩٩٥.

وقد أظهرت النتائج المتحصل عليها أن أغلبية العينات المختبرة تحتوي على متبقيات لمبيدات ألفا وبيتا هكساكلوروسيكلو هكسان، اللندان والد.د.ت ولكن بمستويات أقل من الحد المسموح به طبقا لمنظمة الأغذية والزراعة.

وقد إحتوت بعض العينات على متبقيات الألدرين، ديالدرين والإندرين وبكميات أقل من الحد المسموح به. ولكن أكتشف مبيد ألفا + بيتا هكساكلوروسيكلو هكسان في عينتان من الخيار بمستويات تفوق الحد المسموح به.

وأكتشفت المبيدات الفوسفورية العضوية (الأكتليك والملاثيون) في معظم عينات الخيار والطماطم ولكن بمستويات أقل من الحد المسموح به.

دوليات العلوم الزراعية بمشتمل

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كلية الزراعة بمشتمل



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المجلد الثالث والثلاثون . العدد الأول